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# The effect of populated upper state on the quantum statistics

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## 1. Introduction

The study of the statistical changes in the properties of light due to non-linear interaction between light and matter through the application of quantum statistics has generated substantial current interest (McNeil and Walls 1974, Simaan 1975, Simaan and London 1975a, 1975b). Simaan (1975) has dealt in some detail with the problem of Stimulated Raman Scattering, (SRS) one of the well known and important non-linear interactions, from a two-level system with special reference to the time-evolution of the probability distribution of pump and Stokes fields. He as well as McNeil and Walls (1974) who have tackled the same problem have however neglected the effect of populating the upper state. This assumption, while providing an easy analytic solution for the time-dependence of joint photon probability distribution of Stokes and pump fields, looks somewhat artificial. The aim of the present paper is to lift this assumption thereby making the solutions physically more realistic and giving an answer to the question of whether and how the statistical nature of the pump and stokes fields is affected by the SRS when both upper and lower states are populated. This paper, per necessity, uses the technique of Laplace transforms when both ascending and descending terms of $\rho_{n,m}$ (the distribution function) occur together. The time-variation of mean Stokes and Pump photon number and the coherence characteristics are also calculated computerially.

The equation of motion for the joint probability distribution is obtained in Section 2. The general analytic solution of the equation is derived by Laplace transform method in Section 3. The presence of multiple are also taken care of. Section 4 is devoted to the enunciation of the results. The time variation of the joint photon probability distribution, mean Stokes and Pump photon number, Coherence properties etc. in the presence of a populated upper state are illustrated.

## 2. The equation of motion

A laser pump field of frequency $\omega_l$ is applied in a cavity containing to a two energy level atomic system with energy separation $\hbar\omega$. In the Raman process an incident laser photon at frequency $\omega_l$ is absorbed and either a Stokes photon or an anti-Stokes photon is emitted at frequency $\omega_S = \omega_L - \omega$ or $\omega_{as} = \omega_L + \omega$

respectively. The present discussion, however, is confined to the Stokes field alone, neglecting the anti-Stokes field which can be justified as follows. It is well known that when the upper state is populated there exists a probability for the occurrence of anti-Stokes raman process. This is smaller than the probability of the Stokes process and requires a phase relationship between the fields involved (Garmire, Pandarese and Townes 1963). In this situation the problem can be easily avoided by tuning the cavity to the Stokes frequency. Further, as given in the text of this paper (discussion following equation 5) the upper state population is assumed smaller than the lower state making the Stokes lines much intense than the anti-Stokes.

The equation of motion of the probability distribution function of the joint Stokes-Pump field can be obtained from physical arguments as in Simaan's (1975), or alternatively they are obtained from the master equation approach (Haken 1970, Agarwal 1970, McNeil and Walls 1974) as follows. Substitution of the explicit interaction operator $0 = a_L a_S^\dagger$, where $a_L$, $a_s$ are the annihilation operators and $a_L^\dagger$, $a_s^\dagger$ are the creation operators of Laser and Stokes modes respectively, in the general master equation yields

$$\frac{\partial\rho}{\partial t} = K\beta_1([a_L a_S^\dagger\rho, a_L^\dagger a_S]+[a_L a_S^\dagger, \rho a_L^\dagger a_S])$$
$$-K\beta_2([a_L a_S^\dagger, a_L^\dagger a_S\rho]+[\rho a_L a_S^\dagger, a_L^\dagger a_S]) \quad (1)$$

where $\beta_1$ and $\beta_2$ are the occupation number of the lower and upper level respectively.

From equation (1) the equation of motion for the matrix element of $\rho$ is obtained as

$$\frac{\partial\rho_{n,m}}{\partial t} = 2K\beta_1(n+1)m\rho_{n+1,m-1}-2K\beta_1 n(m+1)\rho_{n,m}$$
$$+2K\beta_2 n(m+1)\rho_{n-1,m+1}-2K\beta_2(n+1)m\rho_{n,m} \quad (2)$$

which can be rewritten as

$$\frac{\partial\rho_{n,m}}{\partial\tau} = (n+1)m\rho_{n+1,m-1}-n(m+1)\rho_n$$
$$+\beta n(m+1)\rho_{n-1,m+1}-\beta(n+1)m\rho_{n,m} \quad (3)$$

where $\tau = 2\beta_1 Kt$ and $\beta = \beta_2/\beta_1$ represents the ratio of the population of the higher to the lower state and $n, m$ represent the pump and Stokes photon respectively.

An observations of equations (2) (or (3)) shows that the first and second terms arise due to Raman process and the third and fourth terms arise to to Inverse Raman process, which are not considered by Simaan (1975) and McNeil and Walls (1974). We neglect the effect of decay on damping for convenience though they can be easily introduced into the calculat on giving only a scaling factor.

### 3. General solution

In order to obtain a general time-dependent solution of equation (3) a Laplace transform method is used. The presence of both ascending and descending terms like $\rho_{n+1,m-1}$ and $\rho_{n-1,m+1}$ renders the calculation more involved and different from the earlier works of Simaan (1975), McNeil and Walls (1974) and others.

A Laplace transform of equation (3) gives

$$\phi_{n,m}(s) = \frac{1}{[s+n(m+1)+\beta(n+1)m]} \times \{\rho_{n,m}(0)+(n+1)m\phi_{n+1,\,m-1}(s)+\beta n(m+1)\phi_{n-1,\,m+1}\} \tag{4}$$

where $\phi_{n,m}(s) = \int_0^\infty \rho_{n,m}(\tau)e^{-s\tau}d\tau$ is the Laplace transform of $\rho_{n,m}(\tau)$. On expanding the R.H.S. of equation (4) it is seen that the number of terms will go on multiplying, increasingly higher powers of $S$ arising in the denominator of subsequent terms. The infinite series thus obtained can be dealt with but a study of theso terms show an well convergent series. So retaining only the first terms, one can write to a good deal of accuracy

$$\begin{aligned}
\phi_{n,m}(s)[s+n(m+1)+\beta(n+1)m] &= \rho_{n,m}(0) \\
&+\sum_{\alpha=1}^{m} \frac{(n+\alpha)!\,m!}{n!\,(m-\alpha)!}\,\frac{\rho_{n+\alpha,\,m-\alpha}(0)}{\prod_{j=0}^{\alpha-1}[s+(n+1+j)(m-j)+\beta(n+2+j)(m-1-j)]} \\
&+\sum_{\alpha=1}^{n} \beta^{\alpha}\,\frac{n!\,(m+\alpha)!}{(n-\alpha)!\,m!}\,\frac{\rho_{n-\alpha,\,m+\alpha}(0)}{\prod_{j=0}^{\alpha-1}[s+(n-1-j)(m+2+j)+\beta(n-j)(m+1+j)]} \\
&+\phi_{n,m}(s)\Bigg[\sum_{j=0}^{m-1}\beta^{j+1}\left(\frac{(n+1+j)!}{n!}\,\frac{m!}{(m-j-1)!}\right)^{2} \\
&\qquad\times\frac{[s+(n+1+j)(m-j)+\beta(n+2+j)(m-1-j)]}{\prod_{k=0}^{j}[s+(n+1+k)(m-k)+\beta(n+2+k)(m-1-k)]^{2}} \\
&+\sum_{j=0}^{n-1}\beta^{j+1}\left(\frac{n!}{(n-1-j)!}\,\frac{(m+1+j)!}{m!}\right)^{2} \\
&\qquad\times\frac{[s+(n-1-j)(m+2+j)+\beta(n-j)(m+1+j)]}{\prod_{k=0}^{j}[s+(n-1-k)(m+2+k)+\beta(n-k)(m+1+k)]^{2}}\Bigg]
\end{aligned} \tag{5}$$

This series is valid for $\beta < 1$ and a similar series can be obtained for $\beta > 1$, though this aspect is not discussed here.

For initial distribution $\rho_{n,m}(0)$ any type of distribution coherent, chaotic or number-state can be chosen. For convenience like others, the initial distribution is taken as $\rho_{n,m}(0) = \delta_{m,0}$. A close observation on the terms of the above series reveals that terms of each series decrease very rapidly so that later terms of the series can be neglected compared to the first ones without much error. Utilizing the above facts and keeping in mind that lower level is more populated than the upper one making $\beta < 1$, after some simplification (5) yields for $m \neq 0$

$$
\begin{aligned}
\phi_{n,m}(s) = {} & \frac{\underline{|n+m}\,\underline{|m}}{\underline{|n}} \cdot \frac{1}{\prod\limits_{j=0}^{m} [s+(n+j)(m+1-j)+\beta(n+1+j)(m-j)]} \\
& \times \Big[ 1 + \frac{\beta(n+1)^2 m^2}{[s+n(m+1)+\beta(n+1)m][s+(n+1)m+\beta(n+2)(m-1)]} \\
& + \frac{\beta n^2 (m+1)^2}{[s+n(m+1)+\beta(n+1)m][s+(n-1)(m+2)+\beta n(m+1)]} \\
& + \frac{\beta^3 (n+1)^2 m^2 (n+2)^4 (m-1)^4}{[s+n(m+1)+\beta(n+1)m][s+(n+1)m+\beta(n+2)(m-1)]^3} \\
& \times \frac{\beta^2 (n+1)^2 m^2 (n+2)^4 (m-1)^4}{[s+(n+2)(m-1)+\beta(n+3)(m-2)]^2} \\
& + \frac{\beta^3 n^2 (m+1)^2 (n-1)^4 (m+2)^4}{[s+n(m+1)+\beta(n+1)m][s+(m-1)(m+2)+\beta n(m+1)]^3} \\
& \times \frac{\beta^2 n^2 (m+1)^2 (n-1)^4 (m+2)^4}{[s+(n-2)(m+3)+\beta(n-1)(m+2)]^2} \Big]
\end{aligned}
\tag{6}
$$

and for $m = 0$

$$
\phi_{n,m}(s) = \frac{\underline{|n+m}\,\underline{|m}}{\underline{|n}} \; \frac{1}{[s+n(m+1)+\beta(n+1)m]} = \frac{1}{s+n} \tag{7}
$$

At this stage it is easy to note that for $\beta = 0$, equation (6) is identical with the corresponding equation of Simaan (1975). The inverse transform of (7) gives

$$
\rho_{n,0}(\tau) = \exp.[-n\tau]. \tag{8}
$$

To obtain the inverse transform of (6) it is observed that all the terms except the first may contain multiple poles for all values of $n$ and $m$ and even the first term will contain a double pole when $m > n$ and

$$j_1+j_2 = (m-n)+\frac{1-\beta}{1+\beta} \tag{9}$$

$j_1$ and $j_2$ being two values of $j$.

However, it is easily noted that since $n$ and $m$ are integers, the above condition (9) will be satisfied when $\beta = 0$ or 1 or when $j_1$ and $j_2$ are non-integers. So the condition will not in general exist in the present discussion. So in the first place an inverse Laplace transform of (6) is obtained when condition (9) does not exist. However, later it is shown that even under condition (9) a solution is possible and in fact such a solution is obtained for small values of $\beta$ in Section 4.

When the first term of equation (6) is devoid of a double pole, the convolution theorem

$$L^{-1}[f(s)g(s)] = [L^{-1}f(s)] * [L^{-1}g(s)] = \int_0^{\tau} f(u)g(\tau-u)du$$

is made use of two take care of the multiple poles in other terms. After repeated application of convolution theorem and using standard Inverse Laplace transform tables (Erdely) *et al* 1954i the inverse transform of (6) and (7) can be written in a general form as

$$\rho_{n,m}(\tau) = A(m, n) \sum_{k=0}^{m} \frac{\exp[-\{(n+k)(m+1-k)+\beta(n+1+k)(m-k)\}\tau]}{\prod\limits_{\substack{j=0\\ j\neq k}}^{m} [(j-k)\{(m-n+1-j-k)-\beta(n+1-m+j+k)\}]}$$

$$+\frac{A(m, n)\beta(n+1)^2m^2}{\{(m-n)+\beta(m-n-2)\}}$$

$$\times \sum_{k=0}^{m} \frac{1}{\prod\limits_{\substack{j=0\\ j\neq k}}^{m} (j-k)\{(m-n+1-j-k)-\beta(n-m+1+j+k)\}}$$

$$\times\left[\exp(-c\tau)\left\{\frac{1-\exp(-d\tau)}{d}\right\}-\exp(-c_1\tau)\left\{\frac{1-\exp(-d_1\tau)}{d_1}\right\}\right]$$

$$+A(m, n)\beta n^2(m+1)^2$$

$$\times \sum_{k=0}^{m+1} \frac{\exp(-c_2\tau)}{\prod\limits_{\substack{j=0\\ j\neq k}}^{m+1} (j-k)\{(m-n+3-j-k)+\beta(m-n+1-j-k)\}}$$

$$\times\left\{\frac{1-\exp(-d_2\tau)}{d_2}\right\}$$

$$+\frac{A(m,n)\beta^3(n+1)^2m^2(n+2)^4(m-1)^4}{\{(m-n)+\beta(m-n-2)\}}$$

$$\times\sum_{k=0}^{m}\frac{1}{\prod_{\substack{j=0\\j\neq k}}^{m}(j-k)\{(m-n+1-j-k)-\beta(n-m+1+j+k)\}}$$

$$\times\left\{\frac{X(a,b,c,d)}{d}-\frac{X(a_1,b_1,c_1,d_1)}{d_1}\right.$$

$$+A(m,n)\beta^3n^2(m+1)^2(n-1)^4(m+2)^4$$

$$\times\sum_{k=0}^{m+1}\frac{X(a_2,b_2,c_2,d_2)}{d_2\prod_{\substack{j=0\\j\neq k}}^{m+1}(j-k)\{(m-n+3-j-k)+\beta(m-n+1-j-k)\}}$$

$$+\delta_{m,0}\exp[-n\tau] \qquad (10)$$

Where

$$A(m,n)=\frac{\lfloor n+m\,\lfloor m}{\lfloor n}(1-\delta_{m,0})$$

$$X(a,b,c,d)=\left[\frac{e^{-a\tau}}{(a-b)^3}\left\{-\frac{a-b}{(a-c)^2}(\overline{a-c}\,\tau+1)-\frac{2}{(a-c)}\right\}\right.$$

$$+\frac{e^{-b\tau}}{(a-b)^3}\left\{-\frac{a-b}{(b-c)^2}(\overline{b-c}\,\tau+1)+\frac{2}{(b-c)}\right.$$

$$+\frac{e^{-c\tau}}{(a-b)^3}\left\{\frac{a-b}{(b-c)^2}-\frac{2}{(b-c}+\frac{a-b}{(a-c)^2}+\frac{2}{(a-c)}\right\}$$

$$\left.-\text{Same terms with } c \text{ replaced by } (c+d)\right]$$

$\delta_{m,o}=1$ for $m=0$
$=0$ for $m\neq 0$
$a=a_1=c_1=[(n+1)m+\beta(n+2)(m-1)]$,
$b=b_1=[(n+2)(m-1)+\beta(n+3)(m-2)]$,
$c=c_2=[n(m+1)+\beta(n+1)m]$,
$a_2=[(n-1)(m+2)+\beta n(m+1)]$,
$b_2=[(n-2)(m+3)+\beta(n-1)(m+2)]$,
$d=[k(m-n+1-k)-\beta k(n-m+k+1)]$,
$d_1=[(k-1)(m-n-k)+\beta(k-1)(m-n-k-2)]$,
$d_2=[(k-1)(m-n-k+2)\beta(k-1)(m-n-k)]$.

## 4. Results and Discussions

The expression for the joint probability distribution of Stokes and pump field (equation 10), as expected, reduces to that of Simaan, when the population of upper level is made zero putting $\beta = 0$. At initial stage for $\tau = 0$, the equation gives $\rho_{n,m} = 0$ for $m$ W 0 and $\rho_{n,m} = 1$ for $m = 0$ which further tends to confirm the correctness of the result.

To illustrate the results a specific case of the incident laser beam with 10 photons is considered. For $\beta = 0{\cdot}1$, the condition (9ĩ can not exist for integral values of $j$ and in that case equation (10ĩ gives the general formula for both $n \geqslant m$ and $n < m$. Figure 1 shows the time-evolution of the joint probability distribution of pump and Stokes photons for $\beta = 0, 1$ and $n+m = 10$. The curve (10, 0), as expected, shows an exponential decrease from the value unity. All other curves except (0, 10) start from zero attains a peak and then gradually decreases, the peak being shifted to higher values of with increase in Stokes photon number, all characteristics as per exception from physical consideration.

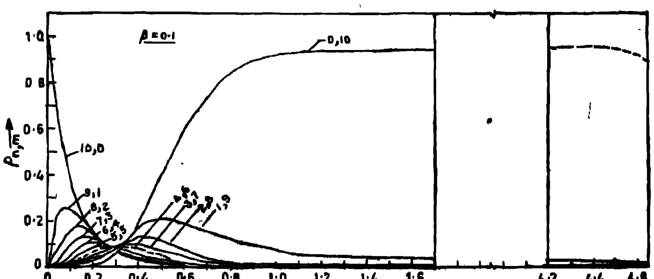


**Figure 1.** Time dependence of the joint photon probability distribution $\rho_{n,m}(\tau)$ with initial distribution $\rho_{10,0}(0)=1$, for the ratio of upper state to lower state population $\beta=0.1$

Unlike Simaan's (1975) time-evolution curves the curve (0, 10ĩ in figure 1, shows a tendency to decrease after a very large time, which is a consequence of including the inverse Raman effect, which will give the correct steady state result. Each of $\rho_{n,m}$'s decreases with increase of $\beta$. This situation for $n = 6$ and $m = 4$ is given in figure 2. For small $\beta$ the results are qualitatively not much different from Simaan's $\rho_{n,m}(\infty)$ increasing with decreasing $n$, making $\rho_{0\,10}$ a maximum and $\rho_{10,0}$ a minimum at large times. The stituation will be reversed for $\beta > 1$.

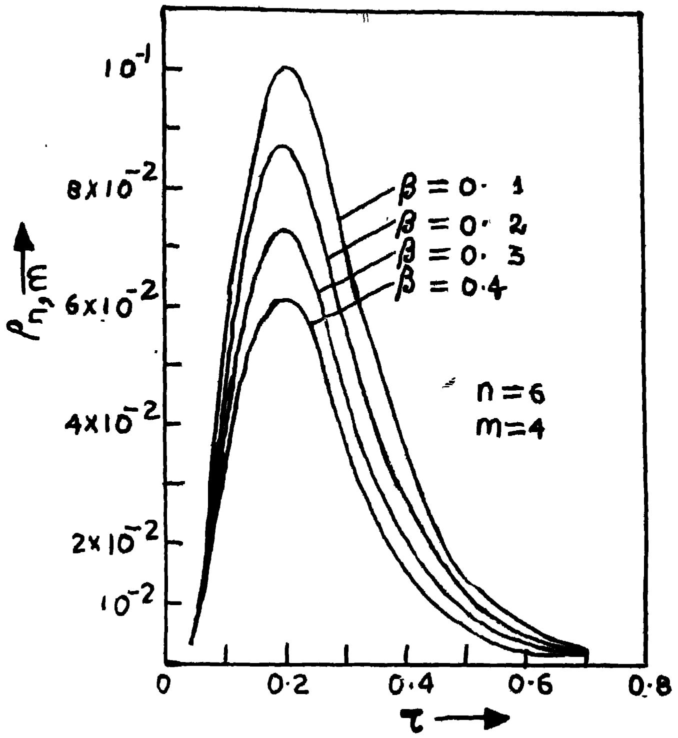


**Figure 2.** Variation of joint photon probability distribution for $n=6$, $m=4$ for different values of $\beta$

If the condition given in equation (9) exists, the double pole occurring in the first term of equation 6 is included and the inverse transform of (6) can be determined as follows.

$$\frac{1}{\prod_{j=0}^{m}[s+(n+j)(m+1-j)+\beta(n+1+j)(m-j)]}$$

is written as equal to

$$\prod_{j_1=0}^{l}[s+(n+j_1)(m+1-j_1)+\beta(n+1+j_1)(m-j_1)]$$

$$\times\frac{1}{\prod_{j_2=l+1}^{m}[s+(n+j_2)(m+1-j_2)+\beta(n+1+j_2)(m-j_2)]}$$

where

$$l = p/2 \quad \text{for even } p$$

$$= \frac{p-1}{2} \quad \text{for odd } p$$

$$p = (m-n) + \frac{1-\beta.}{1+\beta}$$

Then by repeated application of convolution Theorem and taking help of standard inverse Laplace transform tables (Erdelyi *et al* 1954) and keeping terms only up to first order in $\beta$, we obtain

$$\rho_{n,m}(\tau) = A(m,n) \sum_{k=0}^{l} \sum_{k'=l+1}^{m} \frac{1}{\prod\limits_{\substack{j=0 \\ j \neq k}}^{l} (f_j - f_k) \prod\limits_{\substack{j'=l+1 \\ j' \neq k'}}^{m} (f_{j'} - f_{k'})}$$

$$\times \left[ \delta_{f_k f_{k'}} \tau \exp(-f_{k'} \tau) + (1 - \delta_{f_k f_{k'}}) \left\{ \frac{\exp(-f_k \tau) - \exp(-f_{k'} \tau)}{(f_k - f_{k'})} \right\} \right]$$

$$+ \frac{A(m,n)\, \beta (n+1)^2 m^2}{\{(m-n) + \beta(m-n-2)\}} \sum_{k=0}^{l} \sum_{k'=l+1} \frac{1}{\prod\limits_{\substack{j=0 \\ j \neq k}}^{l} (f_j - f_k) \prod\limits_{\substack{j'=l+1 \\ j' \neq k'}}^{m} (f_{j'} - f_{k'})}$$

$$\times \left[ \delta_{f_k f_{k'}} \frac{\exp(-f_1 \tau)}{(f_{k'} - f_1)^2} \{1 - (f_{k'} - f_1)\tau \exp\{-(f_{k'} - f_1)\tau\} - \exp\{-(f_{k'} - f_1)\tau\} \right.$$

$$- \delta_{f_k f_{k'}} \frac{\exp(-f_0 \tau)}{(f_{k'} - f_0)^2} \{1 - (f_{k'} - f_0)\tau \exp\{-(f_{k'} - f_0)\tau\} - \exp\{-(f_{k'} - f_0)\tau\}\}$$

$$+ (1 - \delta_{f_k f_{k'}}) \frac{1}{(f_k - f_{k'})} \left\{ \frac{\exp(-f_1 \tau) - \exp(-f_k \tau)}{(f_k - f_1)} \right.$$

$$- \frac{\exp(-f_0 \tau) - \exp(-f_k \tau)}{(f_k - f_0)}$$

$$\left. \left. - \frac{\exp(-f_1 \tau) - \exp(-f_{k'} \tau)}{(f_{k'} - f_1)} + \frac{\exp(-f_0 \tau) - \exp(-f_{k'} \tau)}{(f_{k'} - f_0)} \right\} \right]$$

$$+ \frac{A(m,n) \beta n^2 (m+1)^2.}{(n-m-2) + \beta(n-m)} \cdot [\text{Same as 2nd term with } f_1 \text{ replaced by } f_{-1}] \tag{11}$$

where

$$A(m, n) = \frac{\underline{|n+m}\ \underline{|m}}{\underline{|n}}$$

$$f_j = [(n+j)(m+1-j)+\beta(n+1+j)(m-j)]$$

$$l = p/2 \qquad \text{for even } p$$

$$= \frac{p-1}{2} \quad \text{for odd } p$$

where

$$p = (m-n)+\frac{1-\beta}{1+\beta}$$

$$\delta_{f_k f_{k'}} = 1 \qquad \text{for } f_k = f_{k'}$$

$$0 \qquad \text{for } f_k \neq f_{k'}$$

The mean laser photon number $<n>$ and the mean Stokes photon number $<m>$ are determined from the relations

$$< n > \; = \; \Sigma \; n\rho_{n,m}$$

and

$$< m > \; = \Sigma \; m\rho_{n,n}$$

since $n$ and $m$ are dependent on each other by the relation $n+m = 10$. For convenience these are determined by a computer calculation and the results are plotted in figure 3, which shows an increase in $<m>$ with decrease in $<n>$ with time.

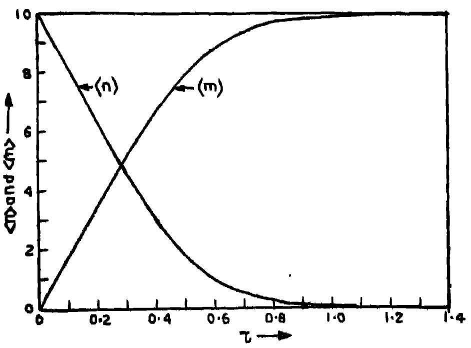


**Figure 3.** Time dependence of mean pump photon number $<n>$ and Stokes photon number $<m>$

Coherence properties of Stokes photons are studied in figure 4 which shows the variation of $\frac{<m^2>-<m>^2}{<m>^2}$ with time. This shows that the outcoming Stokes beam tends to be a nearly coherent after some time. Thus it confirms that from a coherent laser beam of a known frequency, by Stimulated Raman Scattering, another coherent beam at a different frequency can be obtained.

In our calculations we have neglected fifth or more power of $\beta$ compared to $\beta$. The error resulting this is within $(0{\cdot}5)^4 \times 100\%$ that is about 6 per cent which con rms the validity of our approximation.

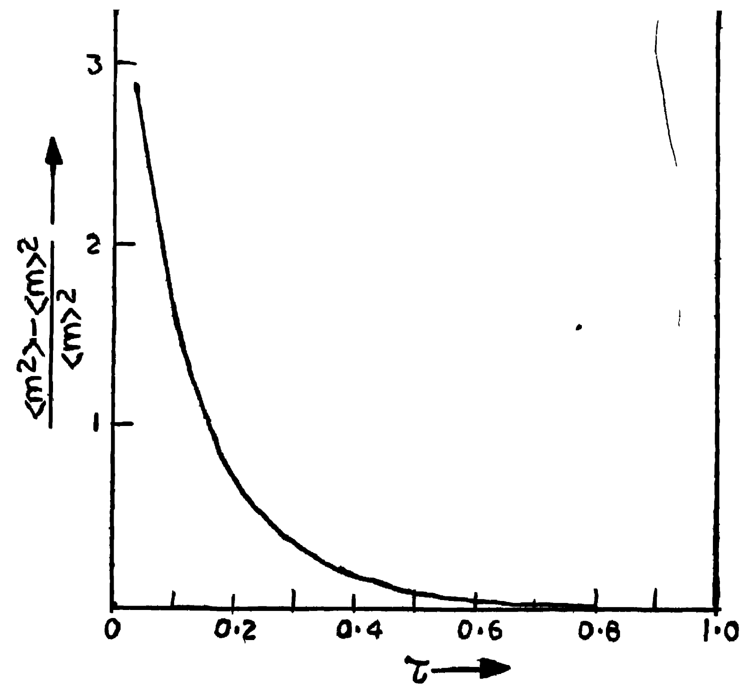


**Figure 4.** Time dependence of Coherence of Stokes photon $\frac{<(m^2)>-<(m)^2>}{<(m)^2}$

One must mention at this point that this paper, as was the case with Simaan's (where the ratio is zero) assumed that the population ratio $\beta$ is a constant, which results in a comparatively simplistic picture of the final Stokes photon conversion. For a rigorous calculation, fluctuation of $\beta$ with time (i.e., higher order non-linear terms in the equation of motion) should be considered and th is is being attempted by us at present.

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